15

THEORY OF SLOW ATOMIC COLLISIONS : OPEN PROBLEMS, NEW TRENDS

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1 INTRODUCTION

This paper is not a review paper on slow atomic collisions. The field is a wide one, many review papers already exist, together with an excellent book written by E.E. Nikitin and S.Ya Umanski (1984). In the framework of the "Séminaire Franco-Roumain", I want to recall some of the main results, and to emphasize the new possibilities opened by the emergence of the cold collisions field.

What do we mean by a "slow" atomic collision ? The collision energy is varying, in the thermal energy range, from a few hundreds down to a few tens of Kelvins (300 K corresponds to 38.7 meV, 10 K to 1.29 meV). The atoms are contained in cells, or moving in atomic beams. They are often excited by laser light. The amount of energy that can be transferred during the collision process is small, only the valence electrons can move from one energy level to another. The inner electrons are not affected by the collision, and can be treated as spectators. Some colliding systems have been widely studied, and can be considered as prototype systems : alkali + rare gas, alkali-earth + rare gas, excited rare gas + ground state rare gas, alkali-alkali systems. The quasi-molecule formed during the collision may then be treated as a one electron or a two electron system.

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MAIN STEPS IN THE EXPERIMENTAL WORK

The developments of the theory are strongly linked to the evolution of the experimental work. We shall recall here below the main steps of this evolution.

Cell experiments

Wood (1914) was the first to observe that a cell containing a mixture of sodium and of rare gas, when irradiated with one of the two lines of the resonance doublet, would reemit the other line. This was the first manifestation of the widely studied fine structure transition :

$$Na(^2P_{1/2}) + X \rightarrow Na(^2P_{3/2}) + X$$

where X is a rare gas. In the 30's, the experiments concerned mainly line broadening and shift and will not be discussed in the present paper. An intense activity in the 60's, connected with optical pumping experiments, and using polarized exciting light, focussed on the study of depolarization, disalignment of a vapor, as well as transfer of population or of orientation between two fine structure levels (Elbel et al, 1972, Krause 1975, Hertel and Stoll, 1977). I want to mention here the experiments of Gay and Schneider (1979 a,b) who studied slow collisions in the presence of a magnetic field. The subject has been revisited recently by a Japanese group (Matsumoto et al, 1991) who observed a magnetic field dependence of the collisional disalignement of neon atoms ($2p^53p$ configuration) in a plasma discharge. This effect could be used for a magnetic field diagnostic. The novelty in the experimental technique is the observation of a time dependent signal following a pulse excitation.

The informations obtained in cell experiments concern total cross-sections, averaged over the velocity distribution and generally summed over some magnetic sublevels. They are not usually very sensitive to the details of the interaction between the two atoms.

Beam experiments

Since 1975, the use of monoenergetic atomic beams, which was first restricted to higher energy collisions, spread into the slow collisions field. Differential cross-sections became available first for alkali rare gas systems (Carter et al, 1975), then for other systems (Colomb de Daunant et al, 1981). Detailed information is then given as a function of the

collision energy and as a function of the scattering angle : the signal usually exhibits interference patterns, so that an accurate comparison between theory and experiment can be performed. From several experiments, it is possible to deduce the interaction potential between the two atoms (see for instance the work of the Pisa group, Aquilanti et al, 1990, and references therein). We should note however that the determination of potentials from laser spectroscopy (Ahmad-Bitar et al, 1977), when it is possible, provides more accurate potentials.

During this period, an intense activity also concerned the study of ionization phenomena :

1) associative ionization discussed in J. Weiner's talk in the present conference (Weiner et al, 1989)

$$A + B \rightarrow AB^+ + e$$

The experiments generally consist in detecting the molecular ion formed during the reaction : detailed results exist in the case of the alkali-alkali systems, as a function of the collision energy and of the polarization of the exciting light . An analysis of the energy spectrum of the emitted electrons (Meijer et al, 1991) provides information on the population of the vibrational state of the product ion.

2)Penning ionization (Hotop, 1980, and references therein, Merz et al, 1990,Müller et al,1991)

$$A^* + B \rightarrow A^+ + B + e$$

The spectroscopy of the emitted electrons provides detailed information on the reaction.

We should note that ionization experiments are very convenient because the detection efficiency of a charged particle is close to one hundred percent.

Cold atoms collisions

Using magneto-optical traps, it is now possible to cool atoms down to a temperature of a few tens of μK, that is to study collisions when the energy is more than six orders of magnitude smaller than in the previous experiments.

The atoms are stored in a given region of the space, so that the information is comparable to what was obtained in cell experiments. However, as the atoms are confined by the laser light, the definition of the cross-section is problematic, the initial and often the final state of the collision being ill-defined. Moreover, the studies as a function of the polarization of the laser light are no longer feasible, since various lasers are involved, propagating in different directions.

The experiments that have been performed up to now are ionization experiments, and concern the same kind of collision partners (alkali + alkali, rare gas + rare gas) as in the previous paragraph. The first experiment concerned the associative ionization reaction :

$$Na + Na + 2h\nu \rightarrow Na_2^+ + e,$$

and was performed by Gould et al (1988). More recently, Lett et al (1991) have shown that the reaction rate depends upon the laser intensity, and presents structures as a function of the laser detuning, demonstrating that the system goes through an intermediate bound molecular state, with vibrational levels.

The Penning ionization reaction between metastable helium atoms has been studied recently (Bardou et al, 1992). The dominant mechanism is apparently :

$$^4He^*(2^3S_1) + {}^4He(2^3P) \rightarrow He + He^+ + e$$

and a cross section of 6 10^6 $Å^2$ is observed, that is much higher than the thermal energy data.

We should note also that beam experiments are performed at temperatures close to 1 mK (Thorsheim et al, 1990)

MAIN STEPS OF THE THEORETICAL WORK

As any collision problem, the theoretical treatment of a slow atomic collision involves two aspects, which should be treated at the same level of accuracy :

1) the potential problem : determination of the interaction between the two atoms at a given internuclear distance R .

2) the dynamical problem : treatment of the relative motion of the two atoms, taking account both of the various potentials and of the dynamical couplings between them.

First step : simple models

Before 1975, the potentials were estimated in the framework of simple models, considering long range polarization forces, and exchange forces. In the latter case, for alkali-rare gas interaction, Fermi (1934) proposed to simulate the rare gas at a distance R by a square well. The excited alkali wavefunction, which is a Rydberg wavefunction, is then phase-shifted due to the presence of this well. The effect of the rare gas can therefore be considered as a R-dependent additional quantum defect : compared to an hydrogenic function, the alkali wavefunction is phase shifted both by the non-hydrogenic potential in the core region, and by the rare gas potential. Later on, such a model has been improved, particularly by the Russian school (Nikitin and Smirnov, 1978), in the framework of the asymptotic methods.

At the same time, simple analytical collision models were proposed, in the spirit of the Landau-Zener model, or of a more elaborate exponential model (Nikitin, 1965, Nikitin and Umanskii, 1980). Such models can provide reliable results for the total cross-sections. Moreover, they are suited to a physical interpretation of the results as a function of well defined parameters.

As a case study, I shall for instance recall the main lines of the mechanisms proposed by Nikitin (1965) for the fine structure transition :

$$Na(3p^2P_{1/2}) + X \rightarrow Na(3p^2P_{3/2}) + X$$

where X is a rare gas. The interaction between the two atoms is anisotropic and depends upon the projection M_L of the sodium atom orbital angular momentum on the internuclear axis. A Σ orbital (M_L=0) is more perturbed than a Π orbital ($|M_L|$=1). This anisotropy is characterized by the quantity :

$$\Delta V(R) = V_\Sigma(R) - V_\Pi(R) \qquad (1)$$

where V_Σ (R) and V_Π (R) are the matrix elements of the alkali-rare gas interaction, treated as a perturbation and diagonal in the subspace of the $|LM_L\ SM_S>$ sublevels of the ^3P term, with L=1 and S=1/2. We shall hereafter write $|M_L, \pm>$, with $M_L = 0, \pm1$, the six substates $|L=1, M_L, S=1/2, M_S = \pm1/2>$. Due to the fine structure effect, the atomic hamiltonian is not diagonal in the $|LM_LSM_S>$representation . The fine structure perturbation $\mathbf{W}= A\ \vec{L}.\vec{S}$ is represented in this subspace by the matrix W_a which contains two identical 1x1 blocks :

$$\Omega = |M_L + M_S| = 3/2 \qquad <1,+|\mathbf{W}|1,+> = <-1,-|\mathbf{W}|-1,-> = \hbar A/2 \tag{2}$$

and two identical 2x2 blocks :

$$\Omega = |M_L + M_S| = 1/2 \begin{pmatrix} -\hbar A/2 & \hbar A\sqrt{2} \\ \hbar A\sqrt{2} & 0 \end{pmatrix} \tag{3}$$

In contrast, the perturbation by the rare gas at a distance R is diagonal in this representation :

$$<1,\pm|\mathbf{V(R)}|1,\pm> = <-1,\pm|\mathbf{V(R)}|-1,\pm> = V_\Pi \tag{4}$$

$$<0,\pm|\mathbf{V(R)}|0,\pm> = V_\Sigma \tag{5}$$

An atomic $|J,M>$ representation with J=3/2, 1/2 can be obtained by diagonalisation of the matrix W_a . The new basis vectors are :

$|3/2,3/2> = |1,+>$ $\qquad$ $|3/2,-3/2> = |-1,->$

$|3/2,1/2> = (\sqrt{2}\,|0,+> + |1,->)/\sqrt{3}$ $\qquad$ $|3/2,-1/2> = (\sqrt{2}\,|0,-> + |-1,+>)/\sqrt{3}$

$|1/2,1/2> = (-|0,+> + \sqrt{2}\,|1,->)/\sqrt{3}$ $\qquad$ $|1/2,-1/2> = (|0,-> - \sqrt{2}\,|-1,+>)/\sqrt{3}$ (6)

In this basis set, the perturbation by the rare gas is represented by a matrix V_c which is not diagonal and contains two 1x1 blocks :

$$<3/2,3/2|\mathbf{V(R)}|3/2,3/2> = V_\Pi \qquad <3/2,-3/2|\mathbf{V(R)}|3/2,-3/2> = V_\Pi \tag{7}$$

and two 2x2 blocks in the subspaces M=1/2 and M=-1/2 :

$$V_c = \begin{pmatrix} \overline{V}+\Delta V/3 & -\sqrt{2}\,\Delta V/3 \\ -\sqrt{2}\,\Delta V/3 & \overline{V} \end{pmatrix} \qquad V_c = \begin{pmatrix} \overline{V}+\Delta V/3 & -\sqrt{2}\,\Delta V/3 \\ -\sqrt{2}\,\Delta V/3 & \overline{V} \end{pmatrix} \tag{8}$$

where $\overline{V}(R) = (1/3)\,V_\Sigma(R) + (2/3)\,V_\Pi(R)$ and $\Delta V(R)$ has been defined in (1) we see that the coupling term is proportional to the *anisotropy* of the perturbing potential.

The adiabatic potential curves may be obtained by diagonalization of the $W_a + V_a$ or $W_c + V_c$ matrices : we obtain the three curves $\Pi_{3/2}$, $\Pi_{1/2}$, and $\Sigma_{1/2}$, :

$$\mathcal{E}(\Pi_{3/2}) = V_\Pi + \hbar A/2 \tag{9}$$

$$\mathcal{E}(\Pi_{1/2}) = \overline{V} + \Delta V/6 - \hbar A/4 + (1/2)\left[(\Delta V)^2 + \hbar A \Delta V + \left[(3/2)\hbar A\right]^2\right]^{1/2}$$

$$\mathcal{E}(\Sigma_{1/2}) = \overline{V} + \Delta V/6 - \hbar A/4 - (1/2)\left[(\Delta V)^2 + \hbar A \Delta V + \left[(3/2)\hbar A\right]^2\right]^{1/2}$$

which are schematically represented in Fig. 1.

At large internuclear distances, the fine structure splitting $\hbar\omega=3\hbar A/2$ is large compared to $\Delta V(R)$: we obtain two eigenvalues, $\hbar A/2$ and - $\hbar$ A, corresponding to the two fine structure levels of the sodium atom : J=3/2 and J=1/2. In contrast, at small internuclear distances the potential anisotropy $\Delta V(R)$ is large compared to the fine structure splitting, so that the electronic hamiltonian is diagonal in the $|LM_L\ SM_S>$ representation., and the molecular potential curves are the V_Σ and V_Π curves. In molecular spectroscopy, the first situation is called " Hund's case c" , because $\Omega= |M_L+ M_S|$ is a good quantum number, while the second situation is called "Hund's case a" because $|M_L|$ and M_S are good quantum numbers . In both cases, the internuclear axis is the quantization axis. A simple explanation for the mechanism of the fine structure transition can be provided by considering a sudden change of coupling from Hund's case c to Hund's case a at the internuclear distance R_0 such that $\Delta V(R_0) = \hbar\omega$. This

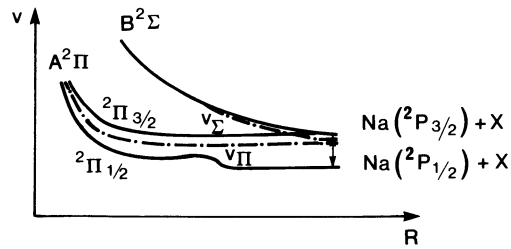

Fig. 1 Molecular adiabatic potential curves for the Na*-X system. The broken line represents the $V_\Sigma(R)$ and $V_\Pi(R)$ curves, which are computed by neglecting the fine structure . For light rare gas perturbers such as He, the $V_\Sigma(R)$ curve is repulsive even at large internuclear distances, whereas it is attractive for heavy perturbers such as Ar.

means that we neglect the electrostatic interaction with the perturber for $R>R_0$, while we neglect the fine structure coupling for $R<R_0$. Let us assume that the relative motion correspond to a rectilinear trajectory, and call $[t_1, t_2]$ the time interval during which $R \leq R_o$. Considering the initial state of the collision to be $|\frac{3}{2}\frac{1}{2}>$, the system at time $t=t_1$ is in the state $|\frac{3}{2}\frac{1}{2}>$. By sudden change of coupling, we obtain (6) a mixture of two states with energy V_Σ and V_Π respectively :

$$t = t_1 \qquad |i> = |\frac{3}{2}\frac{1}{2}> = (\sqrt{2}\ |\ 0,\ +\ >\ +\ |\ 1,\ -\ >)/\sqrt{3} \qquad (10)$$

In the region $R < R_o$, those two states are assumed to be uncoupled and to evolve independently as a function of time so that for $t = t_2$ the system is found in the state $|f>$:

$$|f> = \left[\sqrt{2}\ |0, +> \exp\left\{ -(i/\hbar) \int_{t_1}^{t_2} V_\Sigma dt \right\} + |1, -> \exp\left\{ -(i/\hbar) \int_{t_1}^{t_2} V_\Pi dt \right\} \right] / \sqrt{3} \qquad (11)$$

At $t = t_2$, assuming a sudden change of coupling from Hund's case a to Hund's case c, we obtain the final state of the collision by writing $|f>$ in the $|JM>$ representation. Due to the phase difference between the Σ and Π components, $|f>$ differs from $|i>$ and is a mixture of $|\frac{3}{2}\ \frac{1}{2}>$ and $|\frac{1}{2}\frac{1}{2}>$. For a given impact parameter $b < R_o$, we obtain a probability for fine structure transition $\frac{3}{2}\frac{1}{2} \rightarrow \frac{1}{2}\frac{1}{2}$:

$$P\left(b, \frac{3}{2} \rightarrow \frac{1}{2}\right) = (4/9) \sin^2 \int_{t_1}^{t_2} (V_\Sigma - V_\Pi)\ dt \qquad (12)$$

while the total cross section is :

$$\sigma\left(\frac{3}{2} \rightarrow \frac{1}{2}\right) = 2\,\Pi \int_0^{R_0} P(b)\ b\ db \qquad (13)$$

We therefore obtain an upper bound $(4/9)\ \Pi\ R_o^2$ for this transition, showing that an important parameter is the distance R_o and indicating how the accuracy on the potential curves calculations has an influence on the cross-section .

This model, however, does not take into account the Coriolis coupling due to the rotation of the internuclear axis. At larger collision velocities, or for lighter systems it is no longer appropriate to consider the projection of the angular momentum **J** on the internuclear axis as a good quantum number. For $R > R_o$, the relevant physical model (Masnou-Seeuws and McCarroll, 1974, Nikitin and Umanski, 1984, Kovalenko et al, 1989) considers an atomic electronic angular momentum precessing around an axis Oz perpendicular to the collision plane (Hund's case e). For $R < R_o$, the sudden change of coupling leads to Hund's case b : the spin momentum goes on precessing around the fixed axis Oz, while the orbital angular momentum is precessing around the rotating internuclear axis OZ. Recoupling at time t_2 therefore yields a transition probability which depends not only upon the phase difference between the Σ and Π components of the wavefunction, but also upon the rotation $(\phi_2 - \phi_1)$ of the internuclear axis .

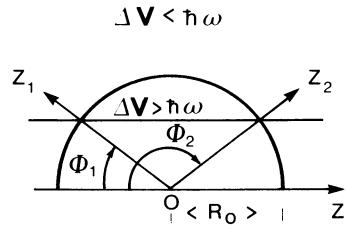

Fig. 2 Rotation of the internuclear axis OZ during the time interval $[t_1, t_2]$ for which the internuclear distance is $R < R_0$, so that $\Delta V(R) > \hbar\omega$.

A good illustration of those coupling schemes, with pictures of the rotating vectors, is given in the paper of Kovalenko et al (1989).

We must note that such simple models provide good agreement with experiment in the case of alkali-rare-gas collisions, and have been confirmed by elaborate semi-classical or quantal calculations (see below). In contrast, for fine structure transitions due to alkali-alkali collisions, the simple model estimations of Dashevskaya (1979) for the cross sections have been confirmed by recent elaborate calculations of Julienne and Vigue (1991), but are in disagreement with the experimental results of Krause and co-workers (Krause, 1975, and references therein). This problem can also be considered as an open problem.

Second step : Elaborate treatment of the dynamics, accurate calculations for the potential curves

Due to the availability of powerful computers in the 70's, the treatment of the dynamics has been markedly improved by numerical solution of the coupled equations for the scattering problem. Both semi-classical methods (Gaussorgues et al. , 1975) with a common trajectory, and quantal methods (Gordon, 1969, Reid and Dalgarno, 1970, Mies, 1973) have been developped, so that it is now possible to obtain the scattering wavefunction with very good numerical accuracy. We should also remark the progress in the description of polarization effects (Nienhuis,1982) .

Between 1975 and 1990, in connection with the availability of accurate experimental spectroscopic data, a huge effort has been undertaken by various groups to obtain reliable potential curves. Indeed, the standard methods of quantum chemistry had been developped with the aim of describing mainly the ground state of the various molecules, in the vicinity of

the equilibrium distance. In contrast, the treatment of a collision problem requires the knowledge of the potential curves over a wide range of internuclear distances, especially at low collision energies where the cross-sections markedly depend upon the long range part of the potential curves. For the interpretation of experiments dealing with laser-excited atoms, one must consider excited and even Rydberg states of the quasi-molecule formed during the collision. For the systems with one or two active electrons, it has been possible to develop specific methods : model potential methods (Dalgarno, 1975, Masnou-Seeuws et al, 1978) or pseudo-potential methods (Pascale, 1983, Müller and Meyer, 1984). The core electrons, moving much more rapidely than the outer electrons, are not included explicitly ; their mean effect on the motion of the outer electron is represented by an effective potential, usually fitted on experimental atomic data. The spirit of such methods is close to the quantum defect approach : for instance, the valence electron of a sodium atom behaves as a Rydberg electron, with an energy-independent quantum defect. The role of the effective potential in the core region, either attractive (model potential) or repulsive (pseudo-potential) is to ensure that outside the Na^+ core the valence electron wavefunction is a Coulomb function with the correct phase shift. The molecular problem is then treated as a one electron-two cores problem (alkali + rare gas, excited rare gas + rare gas) or as a two electron-two cores problem.

In most cases the potential problem can nowadays be considered as solved, at least for short and intermediate ($R < 15\ a_o$) internuclear distances. Before 1980, a 0.1 meV accurary became available for the alkali-rare gas potential curves (Masnou-Seeuws et al, 1978, Pascale, 1983). Next the potential curves for the Ne*-rare gas and Ar*-rare gas systems were computed (see Kucal et al, 1990), and checked by many experiments of the Eindhoven group (Manders et al, 1986). In this case especially for symmetrical systems, an open problem is the treatment of the core-core interaction, which is not addressed by the effective potential methods and requires *ab initio* calculations. For the Penning ionization of the He*-He* system very accurate doubly-excited autoionizing curves have been computed (Müller et al, 1991), in the framework of elaborate *ab initio* calculations : the accuracy on the asymptotes is 5 to 10 meV. Finally, for the alkali-alkali systems, there is now a good agreement between various approaches : we present in fig.3 a recent comparison between two different calculations for excited states of the Na_2 molecule (Magnier et al, 1993). The application of these potential calculations to the theoretical treatment of the associative ionization reaction between two excited sodium atoms is described in O. Dulieu's paper in the present book .

Future work: cold atoms collisions

Turning now to the interpretation of experiments involving cold atoms, we find that the collision problem may be split into two different regions :

a) an inner region ($R < 15\ a_0$) where the moving atoms have been accelerated by the attractive potentials, so that the problem is close to the thermal energy problem. However the number of partial waves is limited due to the centrifugal barrier in the outer region, so that some new effects may appear.

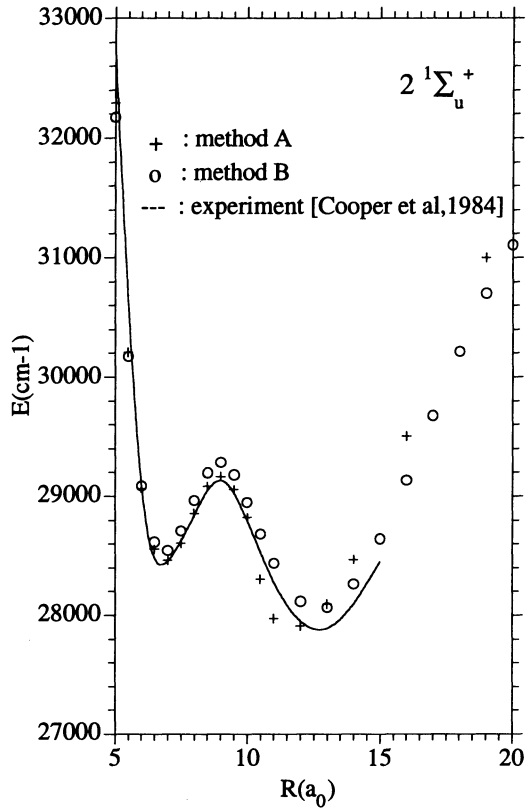

Fig. 3 Extracted from Magnier et al, 1993. Comparison between *ab initio* pseudo potential calculations (method A), model potential calculations (method B), and experiment (solid line) for an excited state of Na_2.

b) an outer region ($R > 15\ a_0$) where the kinetic energy of the relative motion is very small, so that the description of the collision is very sensitive to the details of the potentials, to the fine structure and hyperfine structure couplings, and to any external field. The collision time is no longer short compared to the radiative lifetime, so that the collision problem can no longer be separated from the radiation problem .

The description of this outer region is presently a challenge for theoreticians, as described in the recent work by Julienne and Vigue (1991). The accuracy on the long range potential curves must be improved . The methods for evaluating long range forces have been developped mainly to compute the interaction of two atoms in their ground state, so that the molecular state of the system is well isolated from the excited states . It is no longer the case when one considers two excited atoms . Several groups are working on this subject (Hadinger et al,1992, Scott et al, 1993).

The treatment of the dynamics must also be improved to take account of the electromagnetic field (Band and Julienne,1992)

As a conclusion, we may say that there is a large amount of highly exciting work to be done in the next few years.

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